Advanced nanostructured materials for pushing light trapping towards the Yablonovitch limit

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Abstract: We give an overview on recent progress in the synthesis, fabrication and integration of advanced nanostructured materials for efficient light trapping in high-efficiency thin-film silicon solar cells.
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Tailoring the interaction between light and matter at the nanoscale has gained tremendous importance in the field of photovoltaics, as absorption of sunlight in solar cells can be enhanced drastically by proper engineering of advanced nanostructured materials. Here we give an overview over three novel approaches, recently developed in our lab, to bring light trapping in thin-film silicon solar cells one step closer to the Yablonovitch limit [1].

1. Nanomoulded transparent zinc oxides

Zinc oxide (ZnO) is currently one of the key functional materials for advanced optoelectronic and photonic applications, due to its high transparency across the solar spectrum, excellent electrical properties, and the possibility to synthesize a rich variety of nanostructures. Its abundance and non-toxicity are important additional criteria in view of a global large-scale deployment of photovoltaics. Approaches that have already been successfully employed to increase light trapping in solar modules on millions of square metres [2] include the growth of ZnO films with randomly oriented pyramids by means of chemical vapour deposition [3-4] and wet etching of crater-like structures into sputtered ZnO films [5]. The pyramidal morphology in particular has demonstrated outstanding light-trapping capabilities and has led to several certified world-record conversion efficiencies [6-7]. Solution-based methods have also been extensively investigated for the synthesis of nanopillar-type ZnO structures [8]. Although all these approaches provide a certain degree of freedom in designing the surface morphology of ZnO films, the basic feature morphology (pyramids, craters or pillars) is dictated by the underlying growth and etch kinetics. We recently reported the development of an elegant nanomoulding method [9], which completely frees ZnO films from morphological constraints imposed by nature, and allows one to transfer or replicate an arbitrary master structure made from an arbitrary (transparent or opaque) master material onto a transparent ZnO electrode (Fig. 1).

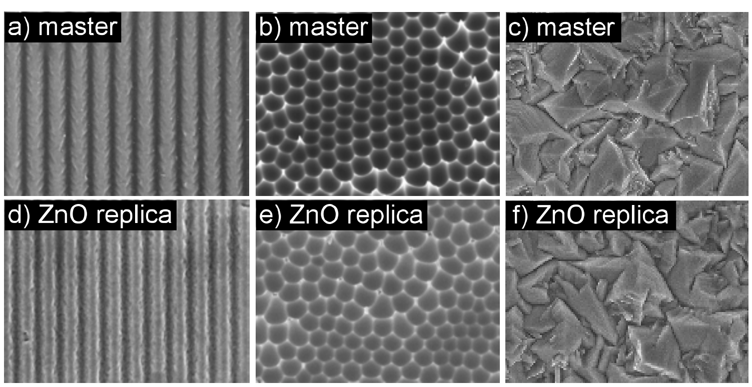

Figure 1: SEM images of master test structures and their corresponding nanomoulded ZnO replicas: a) A one-dimensional periodic grating fabricated by interference lithography, b) a nanotextured aluminium surface with a quasi-periodic hexagonal dimple pattern prepared by anodic oxidation of aluminum, c) random pyramid network of ZnO grown by low-pressure chemical vapor deposition, d-f) images of the corresponding nanomoulded ZnO replica. Image size 6 μm x 4 μm.

We further demonstrated conversion efficiencies for thin-film silicon solar cells deposited on nanomoulded electrodes as high as those on state-of-the-art nanotextured electrodes, proving that ZnO nanomoulding allows one to go far beyond proof-of-concept devices. Nanomoulding therefore provides a promising experimental platform for exploring the light trapping performance of specifically designed photonic nanostructures directly in high-efficiency solar cells.

2. Nanoimprinted superstrates with high-mobility transparent indium oxide

Although a huge potential for boosting the performance of thin-film silicon solar cells has been attributed to nanoimprinting for some time, a clear demonstration in terms of conversion efficiency has been lacking. By replicating the pyramidal morphology of state-of-the-art nanotextured ZnO front electrodes (see Fig. 2), we recently demonstrated excellent conversion efficiencies of 12.0% for amorphous/microcrystalline silicon tandem solar cells. This value, as high as for cells deposited on state-of-the-art ZnO electrodes, validates nanoimprinting as a versatile tool to investigate nanophotonic effects of a large variety of nanostructures directly on device performance [10-11].

Nanoimprinting is performed into a deformable UV-curable lacquer [12], which is both transparent and insulating. An additional transparent conductive layer is therefore required to obtain an operational front electrode. Recently Koida et al. reported the development of a hydrogenated indium oxide (In_2O_3:H) with extraordinarily high mobilities (above 100 $cm^2/V{\cdot}s$) at low carrier densities (in the low 10^{20} cm^{-3} range) resulting in excellent near-infrared transparency [13]. Such high mobilities are obtained after a solid-phase crystallization process at 200° C, which transforms the as-deposited amorphous indium oxide film into a film with small indium oxide nanocrystals, a few hundred nanometers in size, embedded in an amorphous indium oxide matrix [14]. It is possibly this amorphous tissue around the grains, which minimizes grain boundary scattering resulting in Hall mobilities almost as high as the optical mobility. Compared to state-of-the-art ZnO front electrodes, the combination of nanoimprinted superstrate with In_2O_3:H not only leads to a substantial current enhancement of 1 mA/cm^2 in the microcrystalline bottom cell due to the improved near-infrared transparency of In_2O_3:H and the excellent light scattering provided by the nanoimprinted pyramidal morphology, but also an increase of 1 mA/cm^2 in the amorphous top cell due to the higher band gap of In_2O_3:H compared to ZnO [15]. These results show that reducing parasitic absorption in the electrodes becomes increasingly important when increasing the light path in the cell as light is lost in each path through the electrode [16].

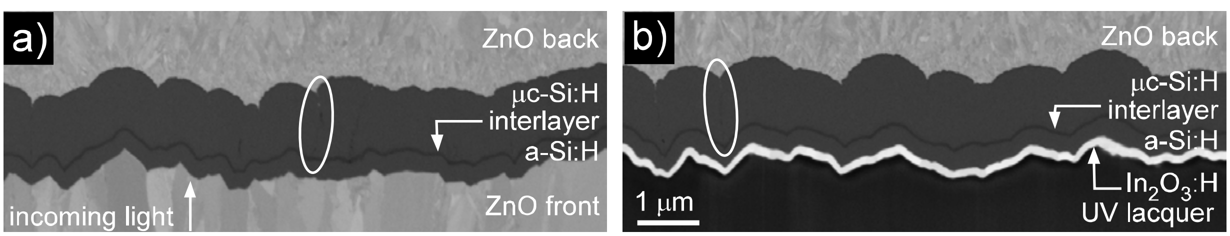

Figure 2: Cross-sections of amorphous/microcrystalline silicon tandem solar cells on a) state-of-the-art randomly textured ZnO front electrode and on b) nanoimprinted replica of ZnO with In_2O_3:H milled by a focused ion beam (FIB) and imaged by secondary electron microscopy (SEM).

3. Nanocrystalline doped silicon/silicon oxide layers

We recently developed novel nanocrystalline mixed-phase silicon/silicon oxide (nc-SiO_x) layers, consisting of nanometric doped silicon filaments embedded in an insulating amorphous silicon oxide matrix. These nc-SiO_x layers allow precise tuning of the optical constants by adjusting the silicon to silicon oxide ratio and are therefore ideal to act as doped silicon window layers [17], to maximize transmission of light into the absorber layer(s). Another important application is their integration as intermediate reflector layer [18] between amorphous top and microcrystalline bottom cell, to boost the current of the top cell (interlayer in Fig. 2). Mixed-phase nc-SiO_x layers have also proven to enable a more tolerant cell design [19], as their low in-plane conductivity limits the negative impact of porous regions (often called cracks marked by ellipses in Fig. 2) in the silicon absorber layer(s) occurring on rough substrates [20], while the high transverse conductivity of the aligned silicon filaments across the layer ensures efficient carrier extraction. Nanocrystalline silicon oxides already clearly demonstrated their potential for enabling solar cells on more aggressive light trapping structures and are therefore instrumental in paving the way for reaching the ultimate limits of light trapping.

4. Conclusion and outlook

We presented various ways of integrating advanced nanostructured materials into high-efficiency thin-film silicon solar cells opening many new opportunities to push light trapping towards the Yablonovitch limit. We demonstrated that nanomoulding and nanoimprinting both enable the fabrication of front electrodes which can rival with state-of-the-art ZnO electrodes, while providing the added flexibility for integrating arbitrary morphologies in high-efficiency solar cells. Future efforts must focus on developing new photonic nanostructures.

We also discussed the importance of reducing parasitic absorption in the cell when the light path is enhanced. In_2O_3:H is a promising new material for front electrodes with minimum parasitic absorption and excellent electrical properties. Parasitic absorption is also an issue in the doped silicon layers and can be minimized by implementing doped nc-SiO_x layers. This material further improves the tolerance of the cell with respect to rough morphologies enabling even more aggressive light trapping schemes in the future.

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